LA-UR-17-30274
Approved for public release; distribution is unlimited.

Title: Nuclear Forensics and Radiochemistry: Cross Sections

Author(s): Rundberg, Robert S.

Intended for: Lecture Series at UC Berkeley

Issued: 2017-11-08

Nuclear Forensics and Radiochemistry: Cross Sections

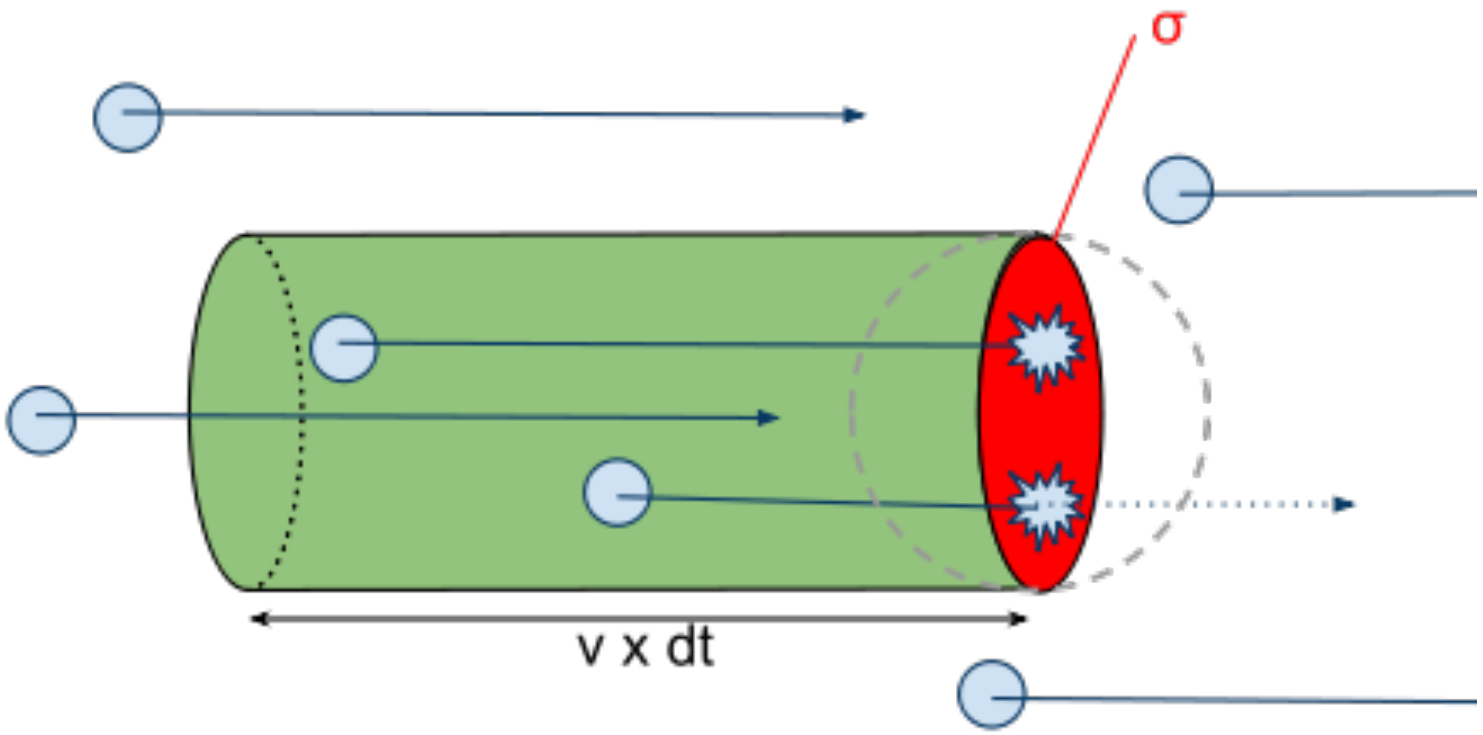
Robert S. Rundberg

Abstract:
The neutron activation of components in a nuclear device can provide useful signatures of weapon design or sophistication. This lecture will cover some of the basics of neutron reaction cross sections. Nuclear reactor cross sections will also be presented to illustrate the complexity of convolving neutron energy spectra with nuclear excitation functions to calculate useful effective reactor cross sections. Deficiencies in the nuclear database will be discussed along with tools available at Los Alamos to provide new neutron cross section data.

Nuclear Forensics and Radiochemistry: Cross Sections

Lecture 4

Simple Concept of Cross Section



Production of an Isotope by Neutron Capture

The rate of neutron capture is,

$$\frac{\mathrm{dN}_{A+1}}{\mathrm{dt}} = \sigma(E)\phi(E,t)N_A.$$

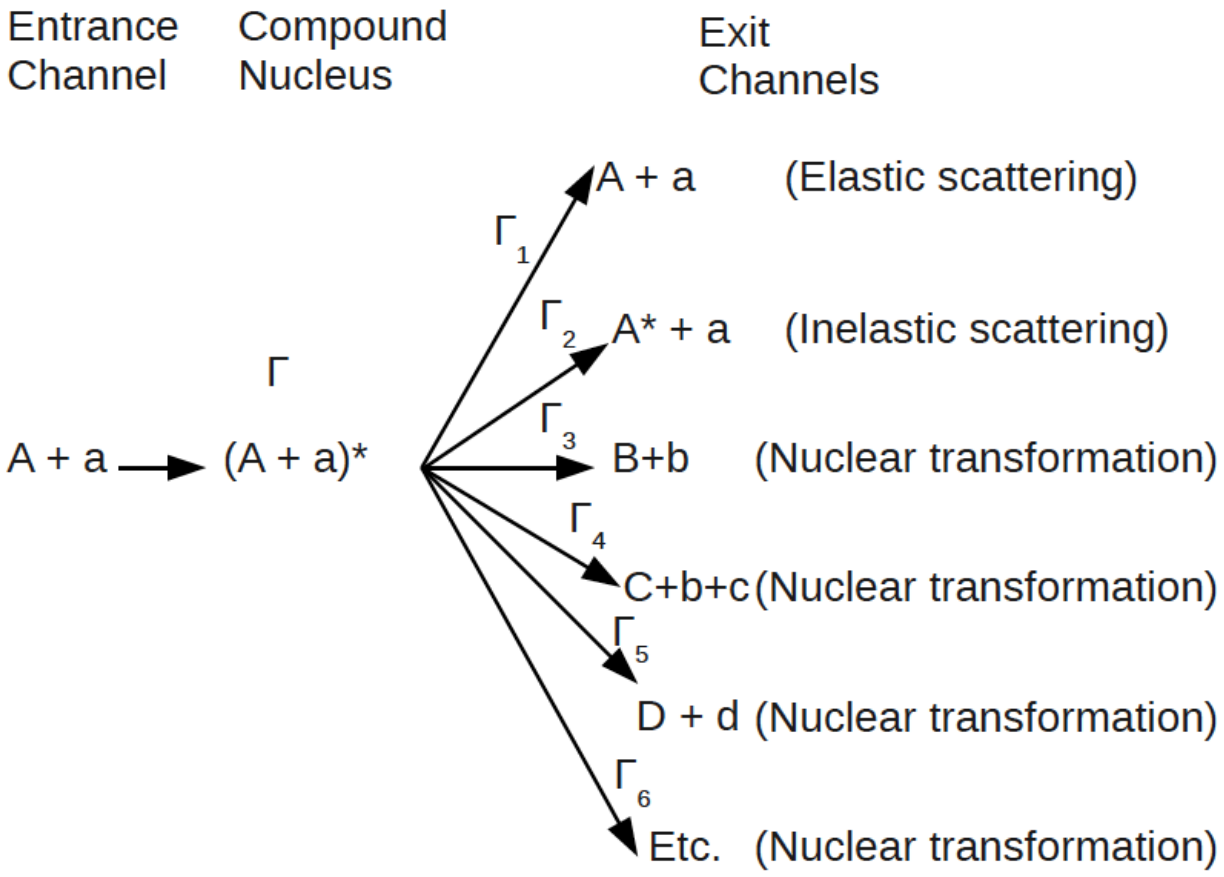
where $\sigma(E)$ is the capture cross section as a function of neutron energy, $\phi(E)$ is the neutron flux as a function of neutron energy, and N_A is the number of target atoms. The number of atoms produced in an experiment is,

$$N_{A+1}(E,t) = \int_0^t \sigma(E)\phi(E,t)N_A \,\mathrm{dt}\,.$$

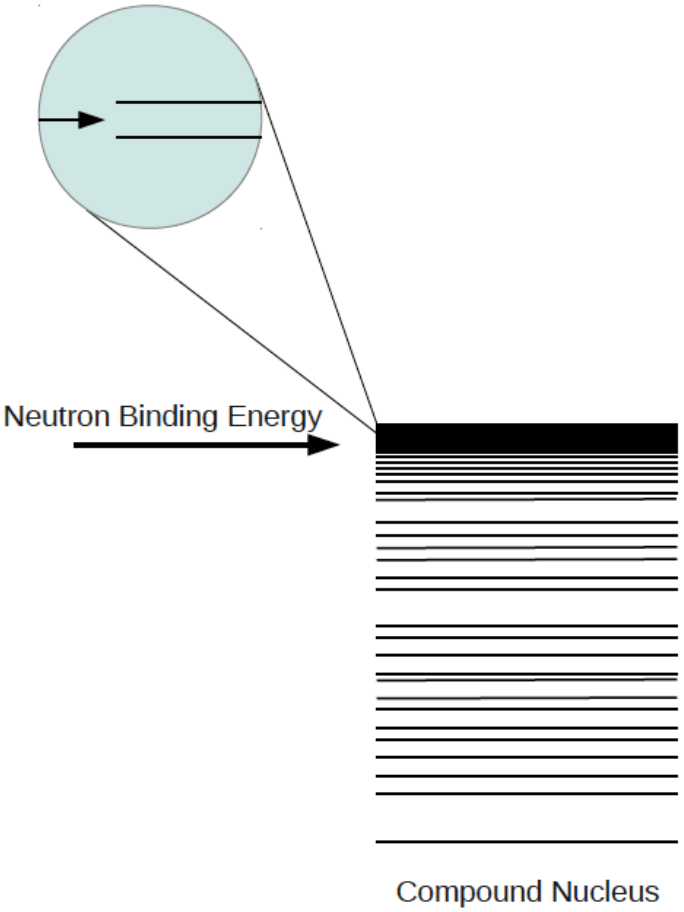
For very fast pulses the integrated flux over time, fluence,$\Phi(E)$ is used and

$$N_{A+1}(E) = \sigma(E)\Phi(E)N_A.$$

The Compound Nucleus



Neutron Resonances are Levels in the Compound Nucleus



Typical Neutron Resonance Spectrum

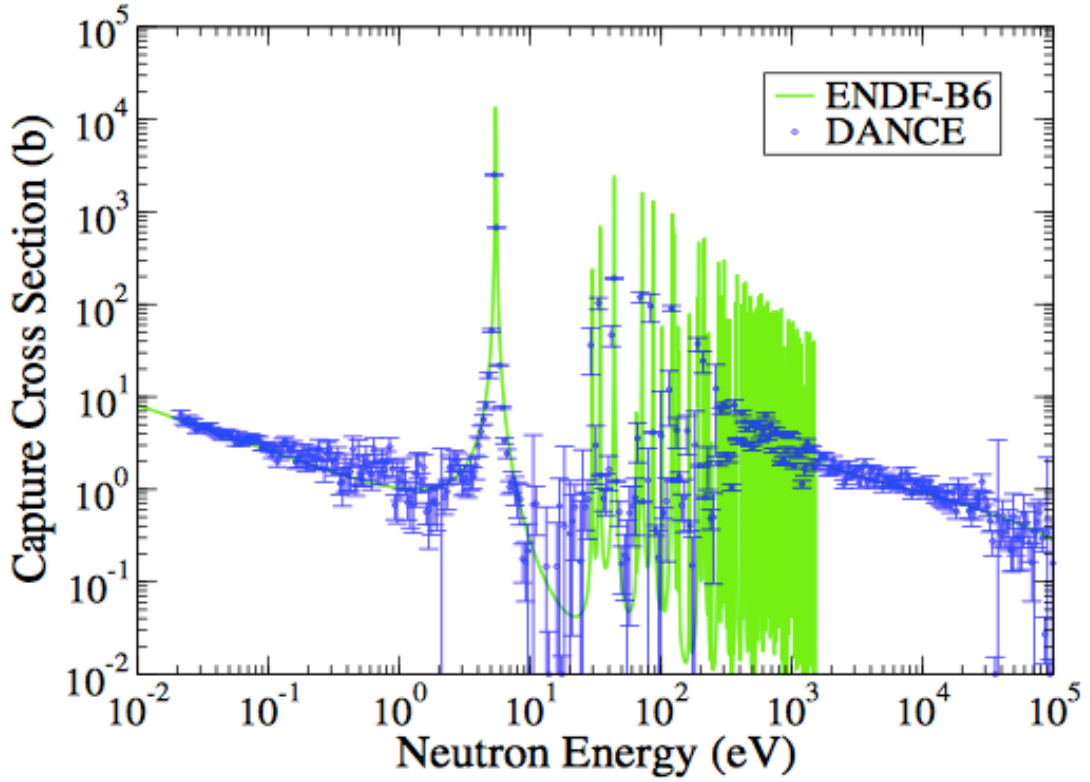


FIGURE 4. ^{236}U capture cross section measured at DANCE.

1/v Cross Section in a Thermal Spectrum is Special

If the neutron velocity distribution is a Maxwell-Boltzmann distribution the flux is,

$$\frac{d\phi(v)}{dv} = 4\pi \left(\frac{m}{2\pi\,\mathrm{kT}}\right)^{3/2} \exp\left(\frac{-mv^2}{2\,\mathrm{kT}}\right) v^2 n_o v.$$

If the neutron reaction cross section has a $1/v$ dependence and the flux distribution does not change with time the number of atoms produced in an event is,

$$N_{A+1} = \int_0^t \int_0^\infty 4\pi \left(\frac{m}{2\pi\,\mathrm{kT}}\right)^{3/2} \exp\left(\frac{-mv^2}{2\,\mathrm{kT}}\right) v^2 n_o \sigma_o \left(\frac{v_o}{v}\right) v dv dt = \sigma_o n_o v_o t.$$

Thus the reaction rate is temperature independent when the cross section has a $1/v$ dependence.

The Breit Wigner Formula

Neutron cross sections have an inherent $1/v$ dependence when the de Broglie wavelength is long compared to the nuclear dimension. This is illustrated in the Breit-Wigner formula for neutron capture to a single state in the compound nucleus,

$$\sigma(n,\gamma) = \pi\bar{\lambda}^2 g \frac{\Gamma_n \Gamma_\gamma}{(E_n - E_o)^2 + (\Gamma/2)^2},$$

where the statistical spin factor $g = \left[\frac{2l_c+1}{(2l_n+1)(2l_t+1)}\right]$, Γ_n is the neutron width, Γ_γ is the gamma width, and Γ the total width for the compound nucleus. as $E_n \to 0$, the Lorentzian becomes a constant $1/E_o^2$. The deBroglie wavelength, $\bar{\lambda} \backsim 1/v$, and $\Gamma_n \backsim v$, thus,

$$\sigma(n,\gamma) \backsim 1/v.$$

The area under a single resonance is,

$$A = 2\pi^2 \bar{\lambda}^2 g \frac{\Gamma_n \Gamma_\gamma}{\Gamma},$$

which is proportional to $1/v$ when $\Gamma_n \ll \Gamma_\gamma$.

Area Under Lorentzian

Area under a Lorentzian: details

To find the area under a Lorentzian (i.e. to integrate from $-\infty$ to $+\infty$), make the following substitution:

$$(\nu - \nu_0) = \gamma \tan\theta$$

(Obviously if the Lorentzian is given with different constants, choose appropriately: you want to end up with just $(1 + \tan^2\theta)$ times some constant in the denominator.) Then,

$$d\nu = \gamma \sec^2\theta = \gamma\,(1 + \tan^2\theta)\,d\theta$$

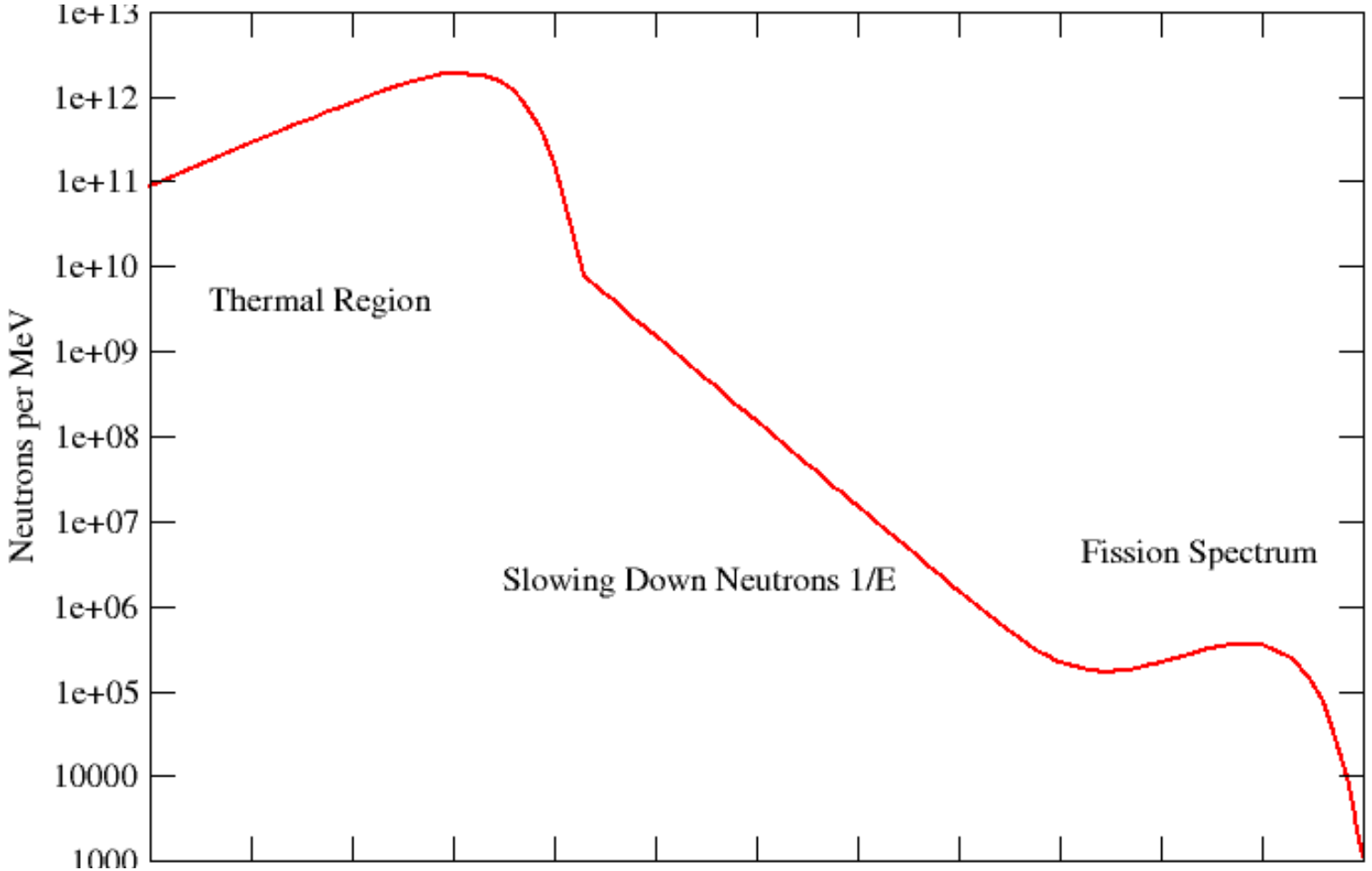
(the latter following from the Pythagoras identity). With the above substitution, the new limits on the integral become $-\pi/2$ and $+\pi/2$ because that's where $\tan\theta$ goes to $-\infty$ and $+\infty$ respectively. So now we have

$$\int_{-\frac{\pi}{2}}^{+\frac{\pi}{2}} \frac{\gamma \cdot \gamma(1 + \tan^2\theta)\,d\theta}{\gamma^2(1 + \tan^2\theta)}$$

which simplifies very nicely to

$$\int_{-\frac{\pi}{2}}^{+\frac{\pi}{2}} d\theta = \left[\left(\frac{\pi}{2}\right) - \left(-\frac{\pi}{2}\right)\right] = \pi$$

Reactor Spectrum



Flux Distributions

Watt Spectrum Phys.Rev. **87**, 1037(1952)

$$P(E) = 0.4865 \sinh(\sqrt{2E}) \exp(-E) \text{ MeV}^{-1} \quad (1)$$

See Nuc. Phys. A **957**, 289(2017) for a more accurate model of fission neutrons.

Maxwell Boltzmann Distrbution

$$f(E) = 2\sqrt{\frac{E}{\pi}} \left(\frac{1}{kT}\right)^{3/2} \exp(-E/kT) \quad (2)$$

Effective Reactor Cross Sections

In the Wescott convention, the neutron reaction rate for a given nuclide is $R_s = n\nu_0\hat{\sigma_0}$, where the effective cross section is given as $\hat{\sigma_0} = \sigma_0(g + rs)$. g is a neutron temperature dependent factor that accounts for the deviation of the excitation function from a $1/v$ dependence. r is the ratio of epithermal neutrons to thermal. s is the ratio of the reduced epithermal integral to thermal. σ_0 is the thermal cross sections defined at 2200 m/s neutron velocity.

s is calculated as

$$s\sqrt{\frac{T_0}{T}} = \frac{2}{\sqrt{\pi}}\frac{\Sigma'}{\sigma_0} \tag{1}$$

where Σ' is the cross section above the cadium cutoff integrated over a $1/E$ spectrum with the $1/v$ cross section subtracted and

$$r\sqrt{\frac{T}{T_0}} = f\sqrt{\pi\mu}/4 \tag{2}$$

where f is the ratio of epithermal flux to the thermal flux, μkT is the energy below which the epithermal neutron spectrum ends.

Westcott-g

The neutron temperature dependent parameter g is defined as the Maxwell Boltzmann average reaction rate over the rate for a $1/\nu$ absorber,

$$g = \frac{\sigma_T}{\sigma_0} = \frac{1}{\sigma_0 \nu_0} \int \frac{4}{\sqrt{\pi}} \left(\frac{\nu}{\nu_t} \right)^3 e^{-(\nu/\nu_t)^2} \sigma(\nu) d\nu. \tag{3}$$

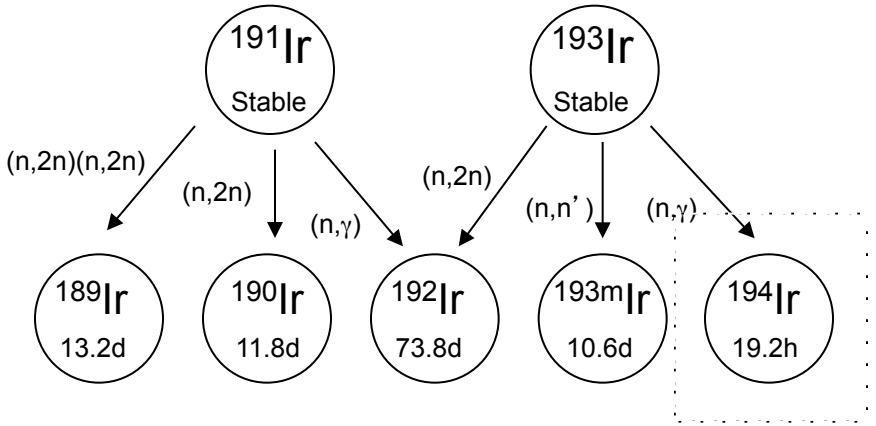
Activation Products

- Neutron activation products can provide forensic information about:
  - Materials used as device components.
  - The location of materials.
  - The neutron spectrum that materials were exposed to.
  - The amount of environmental material swept up into the fireball.
- Neutron activation of ubiquitous materials may aid in the determination of nuclear yield.
- Neutron capture reactions are sensitive to the details of the low energy spectrum.

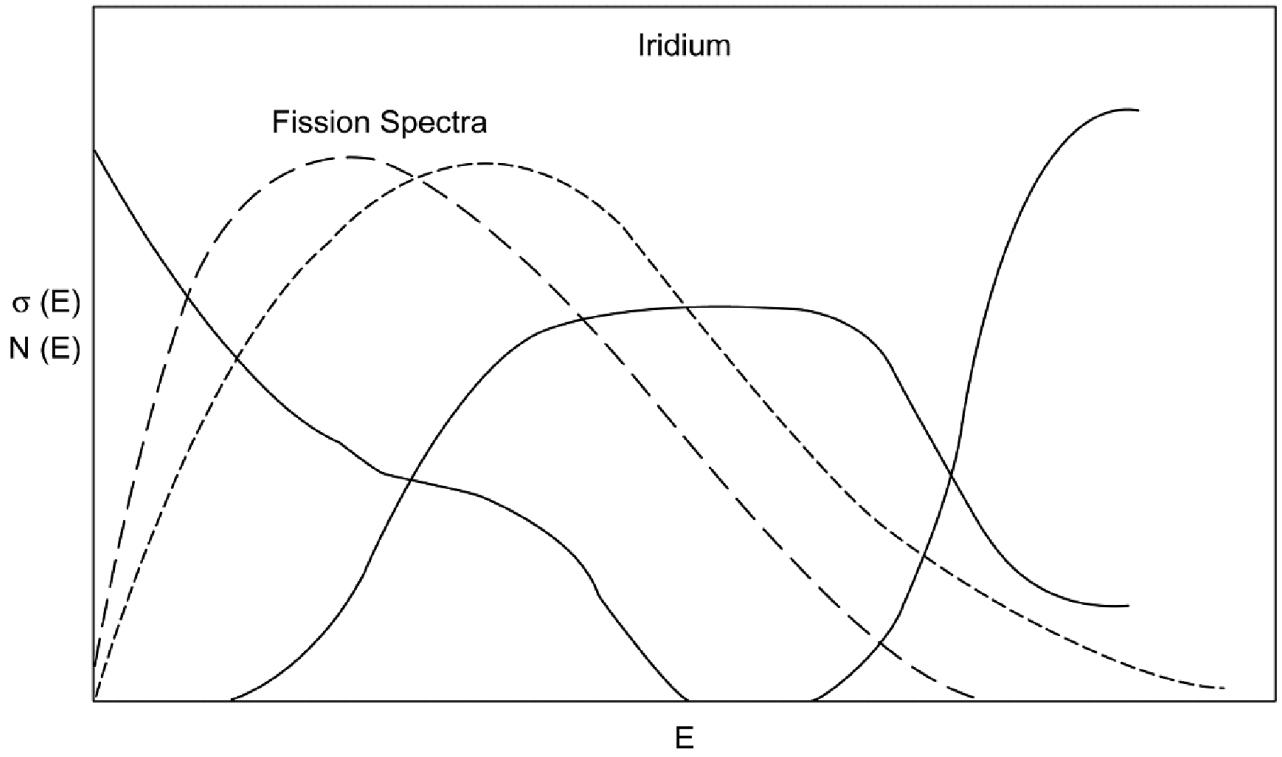
Iridium Activation

Gives information about:

- Neutron energy > 8MeV
  - $^{191}Ir(n,2n)^{190}Ir$
- Neutron energy > 1 MeV
  - $^{193}Ir(n, n')^{193m}Ir$
- Low energy neutrons
  - $^{191}Ir(n,\gamma)^{192}Ir$
- Fast flux
  - $^{191}Ir(n,2n)^{2\ 189}Ir$



Iridium Excitation Functions



Nuclear Decay Characteristics for Ir Isotopes

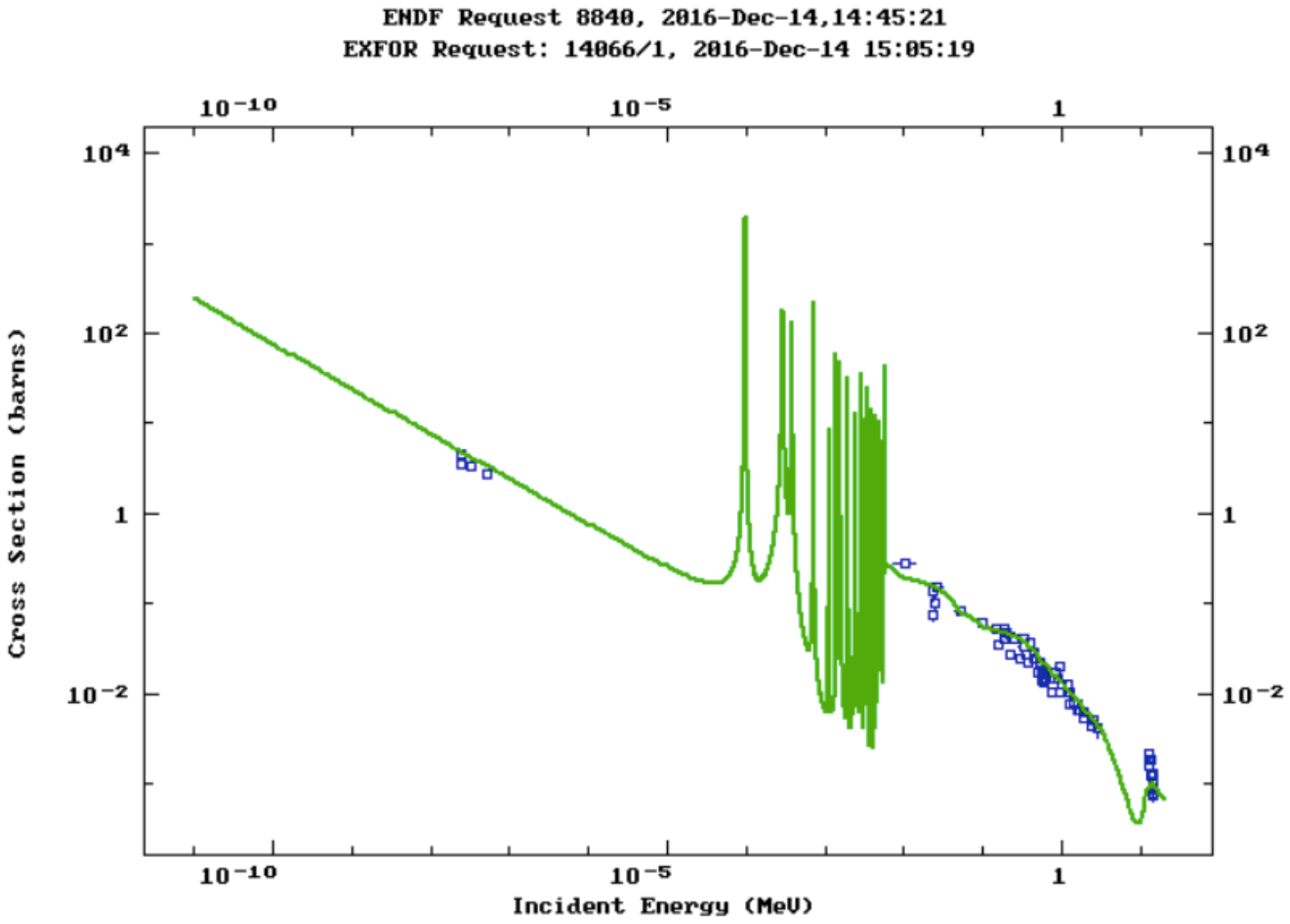
Isotope	Produced by	Half-Life	Mode of Decay	Gamma Radiations (MeV)	x-Rays
^{189}Ir	$(n,2n)^2$	13.10 d	EC	0.245	Os K and L x-rays
^{190}Ir	(n,2n)	11.78 d	EC	0.186, 0.518, 0.605	Os K and L x-rays
^{192}Ir	(n,γ) and (n,2n)	74.2 d	β-, EC	0.308, 316, 468	Pt K and L x-rays Os K and L x-rays
^{193m}Ir	(n,n')	10.60 d	IT		Ir K and L x-rays
^{194}Ir	(n,γ)	19.15 h	β-	0.294, 0.328	

X-ray Energies (keV)

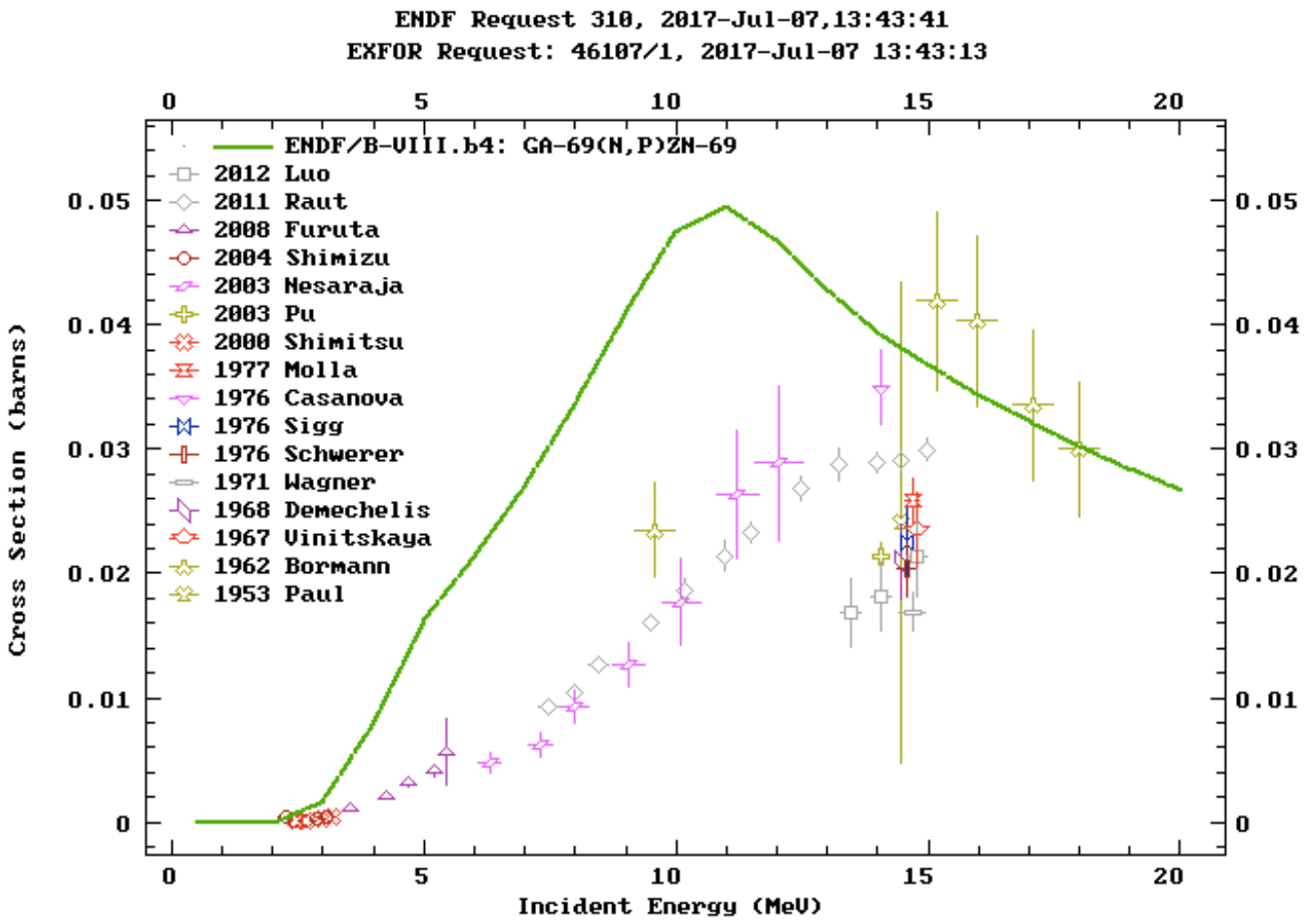
Element	K_α	K_β	L_α	L_β	L_γ
Os	61.48> 62.99	71.40> 73.30	8.84 8.91	10.35 10.60	12.09
Ir	63.28> 64.89	73.55 75.61	9.10 9.17	10.71 10.92	12.51
Pt	65.11 66.82>	75.74 77.87>	9.36 9.44	11.07 11.25	12.94

Nuclear decay characteristics for ^{189}Ir, ^{190}Ir, ^{192}Ir, ^{193m}Ir, and ^{194}Ir.

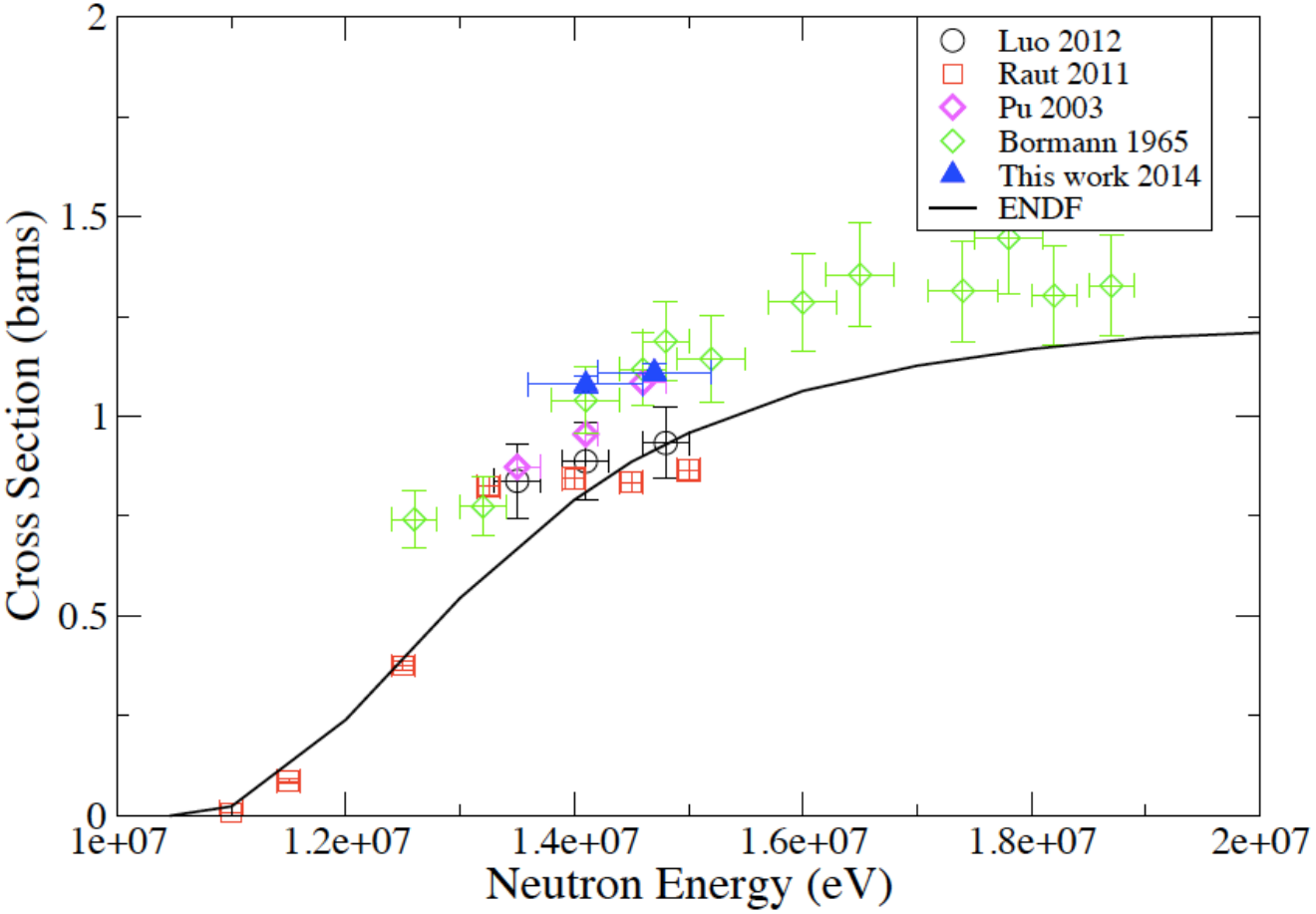
Gallium Cross Section Needs



Another Gallium Cross Section Need



Mistakes Have Been Made

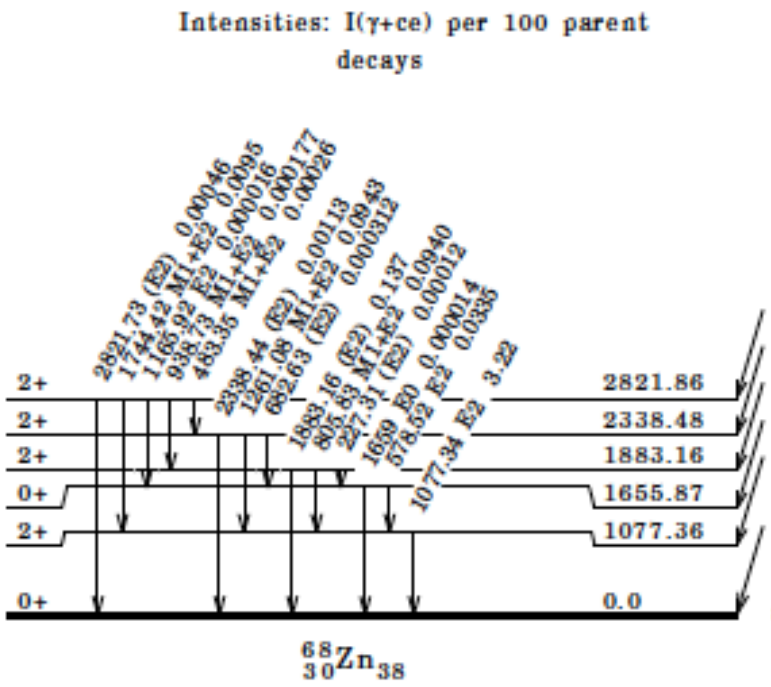


Ga-68 Decay Scheme

^{68}Ga ε Decay 1999BeZQ,1999BeZS

Decay Scheme

Intensities: I(γ+ce) per 100 parent decays



Eε	Iβ+	Iε	Log *ft*
		0.0104	5.113
		0.0955	5.754
		0.230	5.879
	0.000258	0.0328	6.898
	1.19	1.80	5.489
2912	87.72	8.92	5.1942

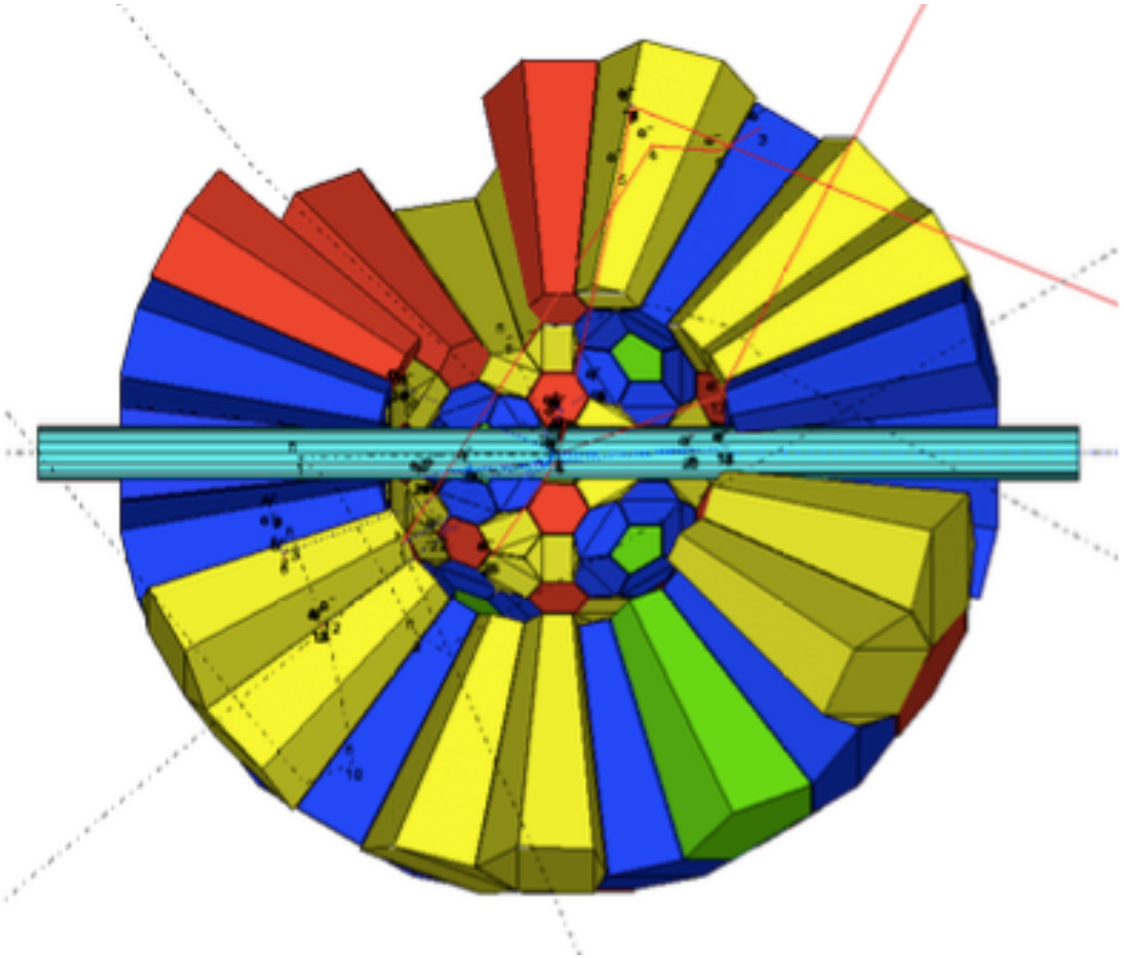
stable

Annihilation Gamma Ray Summing

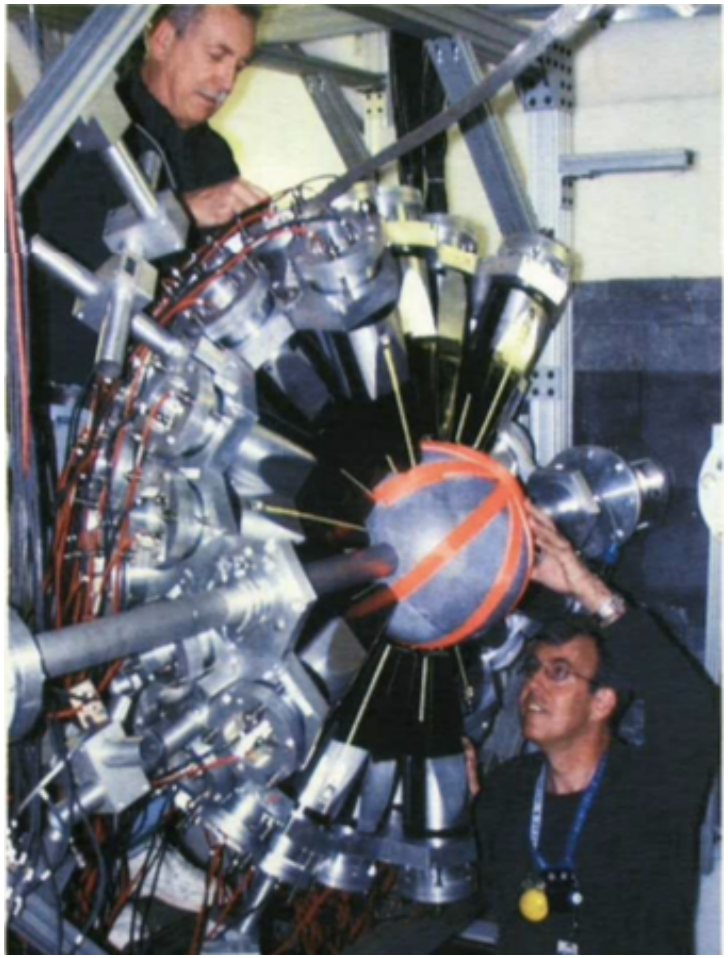
$$R = \frac{f_1 \varepsilon_1^P}{1+\alpha_1}\left(1 - 2\frac{f_2 \varepsilon_2^T}{1+\alpha_2}\right)A$$

Summing with annihilation radiation is a special case because the photons are spatially correlated. The probability of detecting a 511 keV photon is doubled.

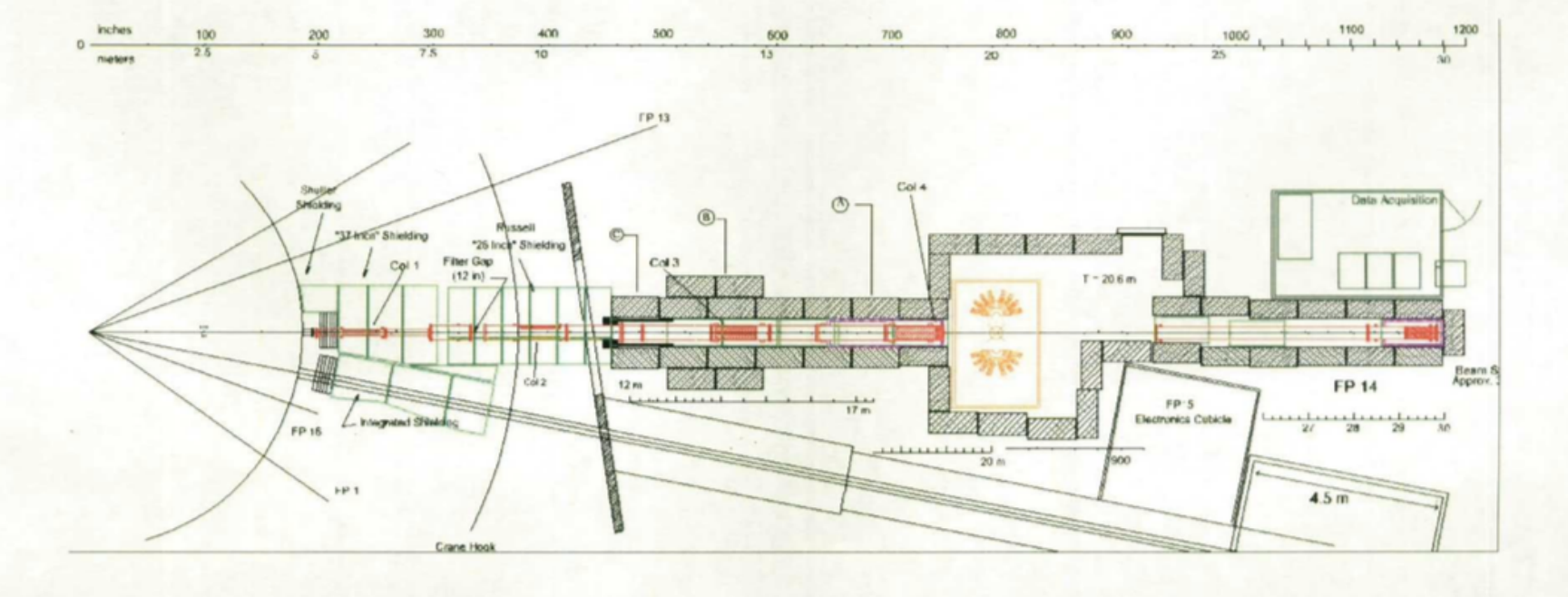
Device for Advanced Neutron Capture Experiments, DANCE



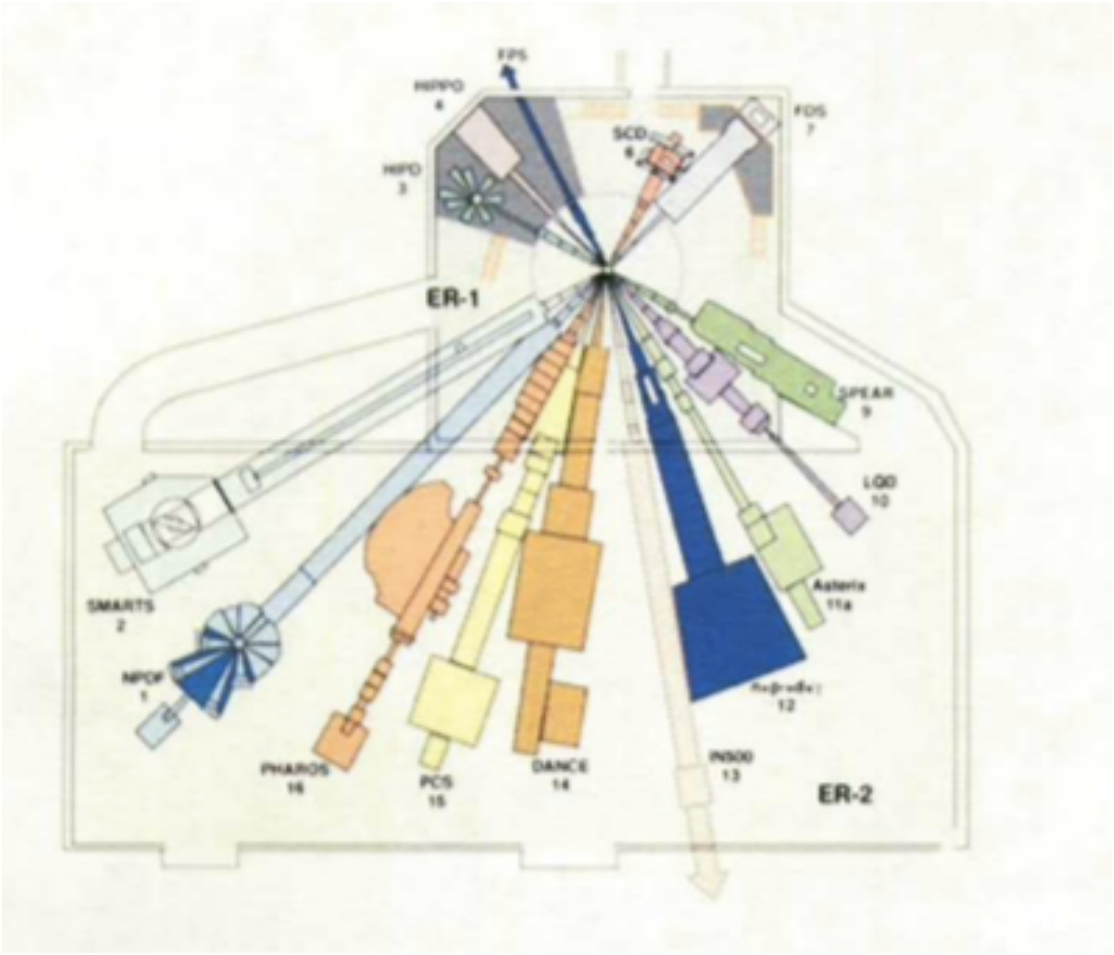
DANCE



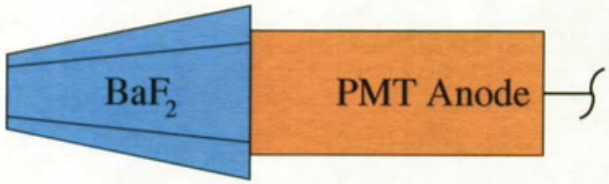
Layout of the DANCE Time-of-Flight Spectrometer



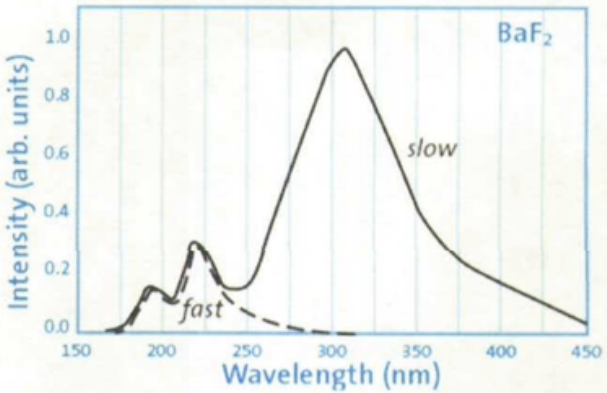
DANCE Location in the Lujan Center

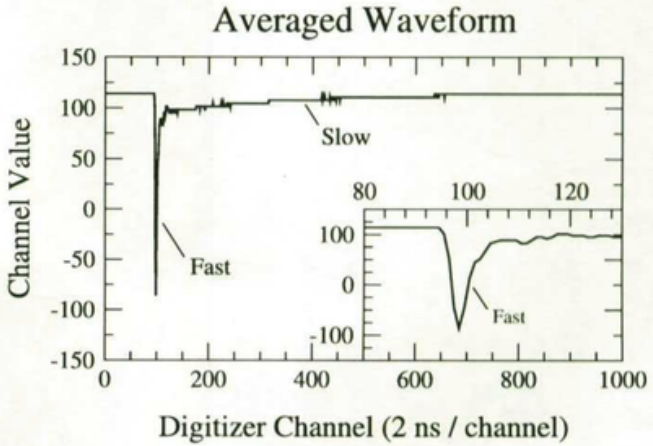


Properties of BaF_2 Scintillators



- The total light output from BaF_2 arises from two scintillation mechanisms, which are differentiated by their respective decay times (*e.g.* fast and slow).
- BaF_2 suffers from an intrinsic α activity (averaging ~ 225 cnt/s/l for crystals in the DANCE array) resulting from unavoidable Ra contamination (a chemical homologue of Ba).



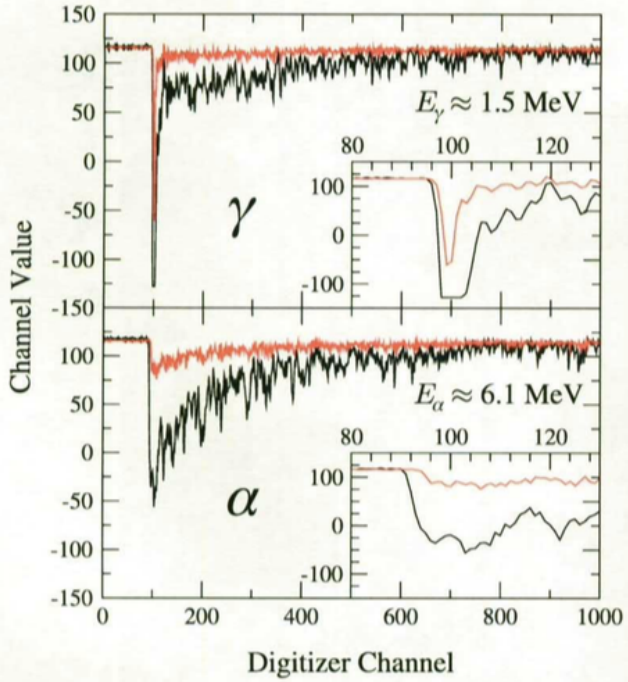


Scintillation Properties

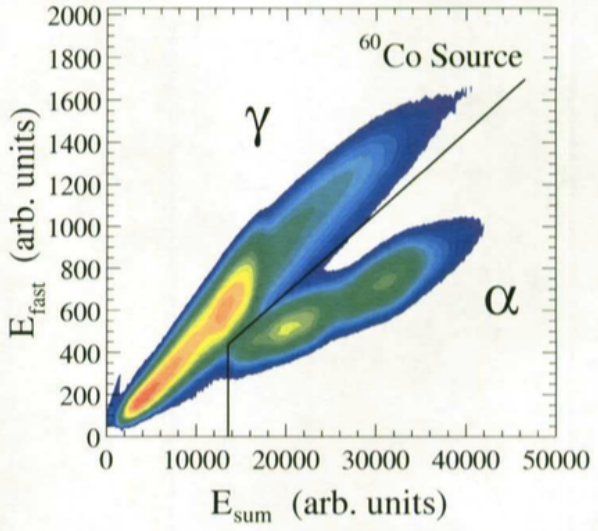
Scintillation Properties	Fast	Slow
Emission Maximum λ (nm)	220	310
Primary Decay Time (ns)	0.6 – 0.8	630
Light Yield (photons/MeV·g)	2.5×10^3	6.5×10^3
Photoelec. Yield (% of NaI(Tl))	5	16

DANCE Pulse Shape Discrimination

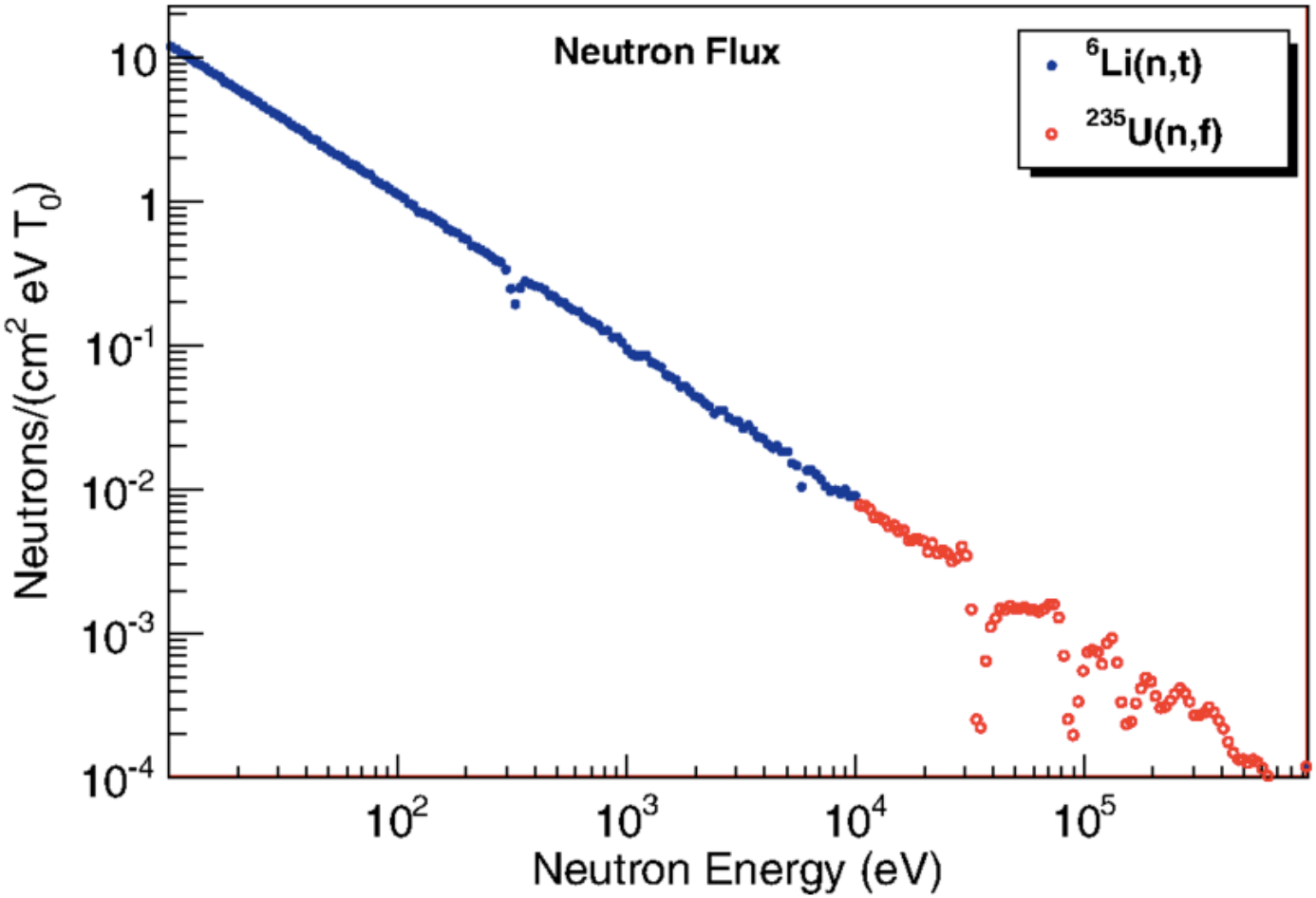
Relative intensities of fast and slow components exhibits a strong dependence on the type of radiation detected.



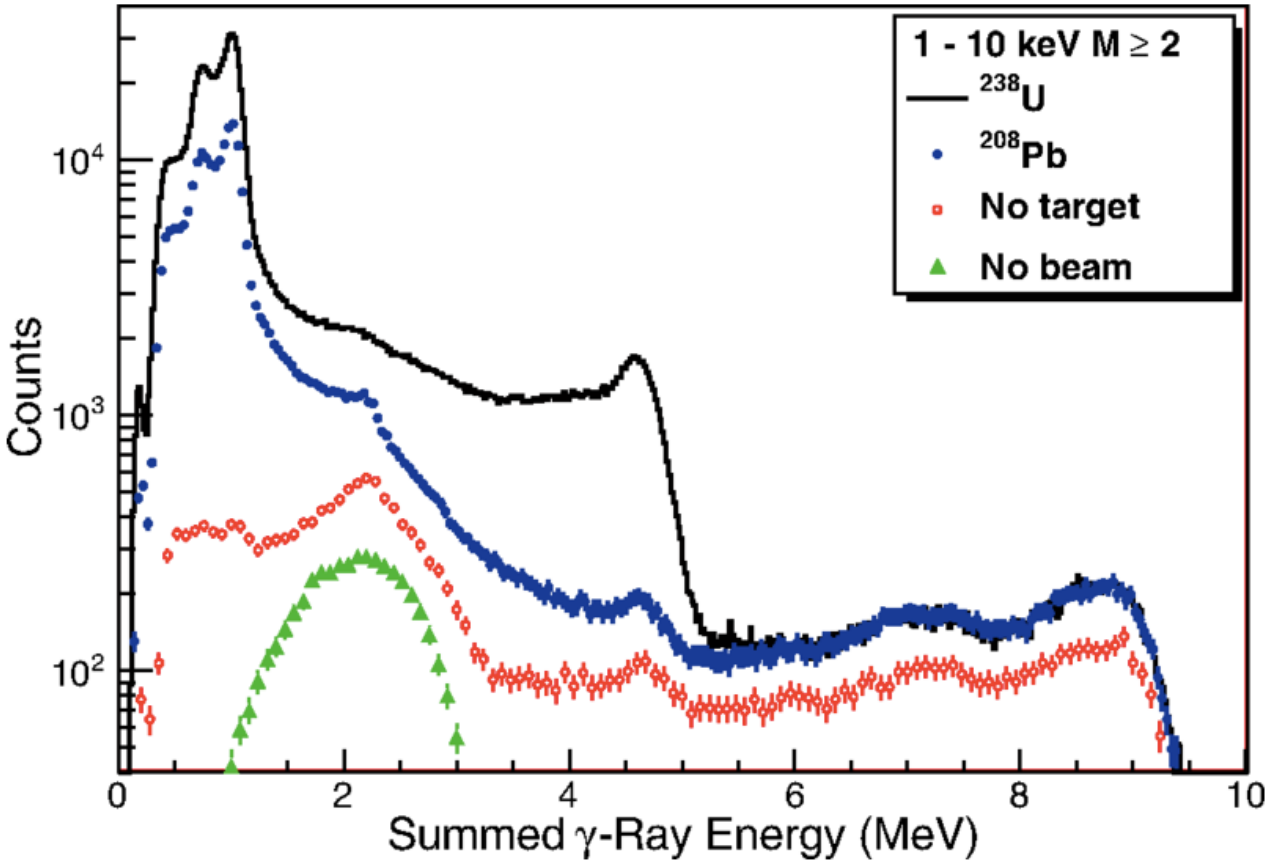
α-particles deposit energy almost exclusively through the slow component mechanism. Comparison of the fast and slow component integrals provides a means to reject the background α activity.



DANCE Neutron Flux

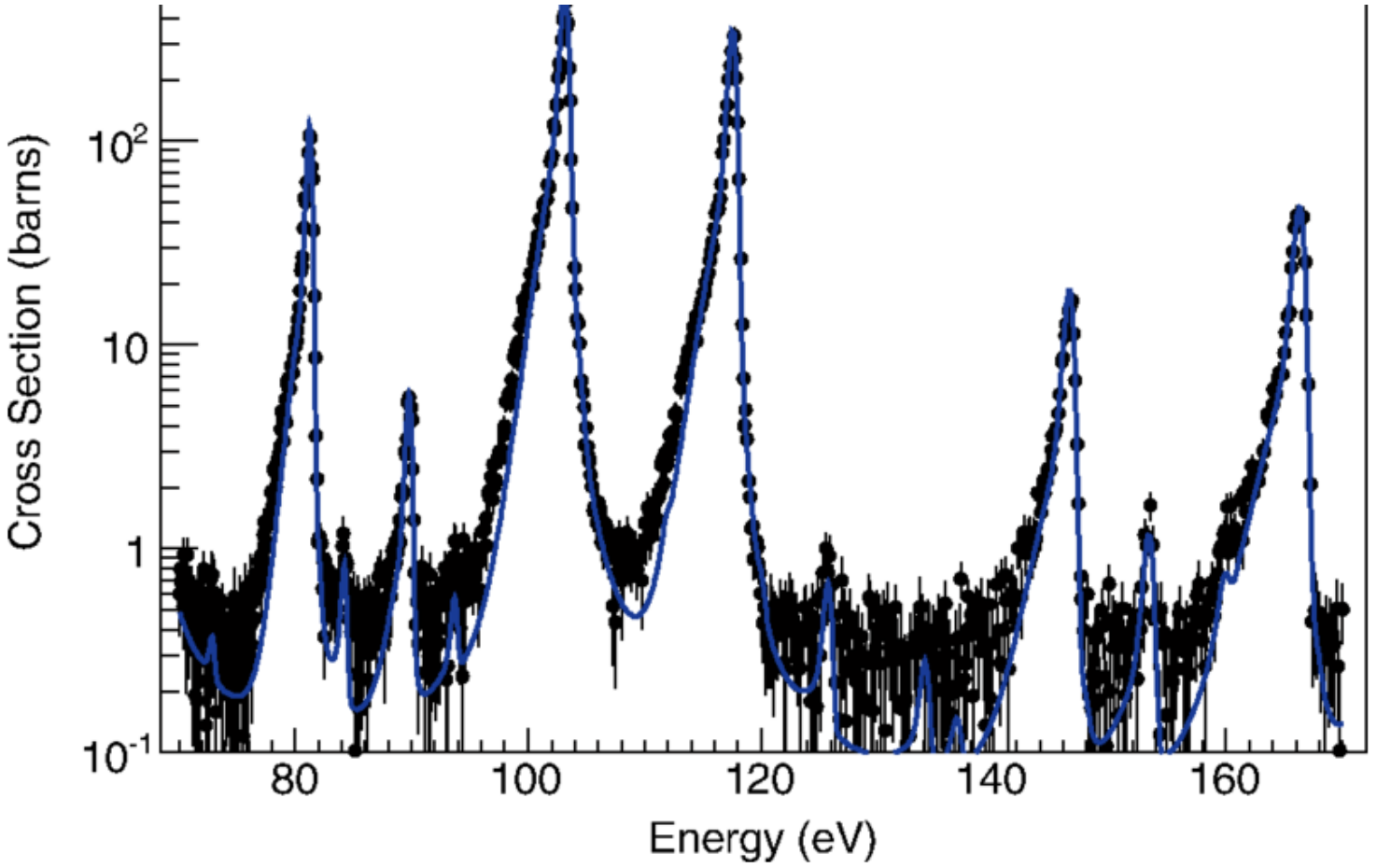


DANCE Backgrounds

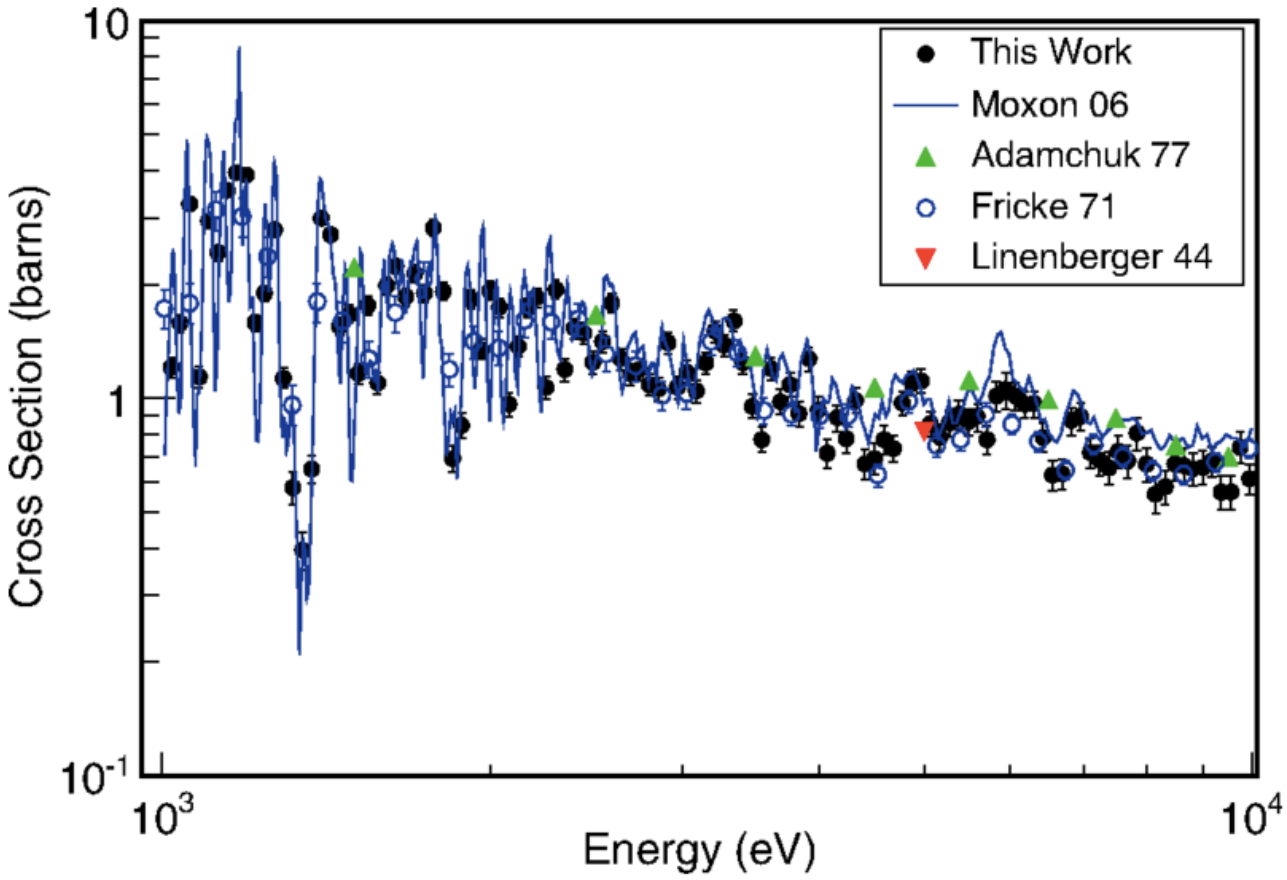


Summed gamma-ray energy spectra for reactions with neutron energies between 1 and 10 keV and gamma-ray multiplicity $M_{cl} \geq 2$.

U-238 n, gamma Resonances

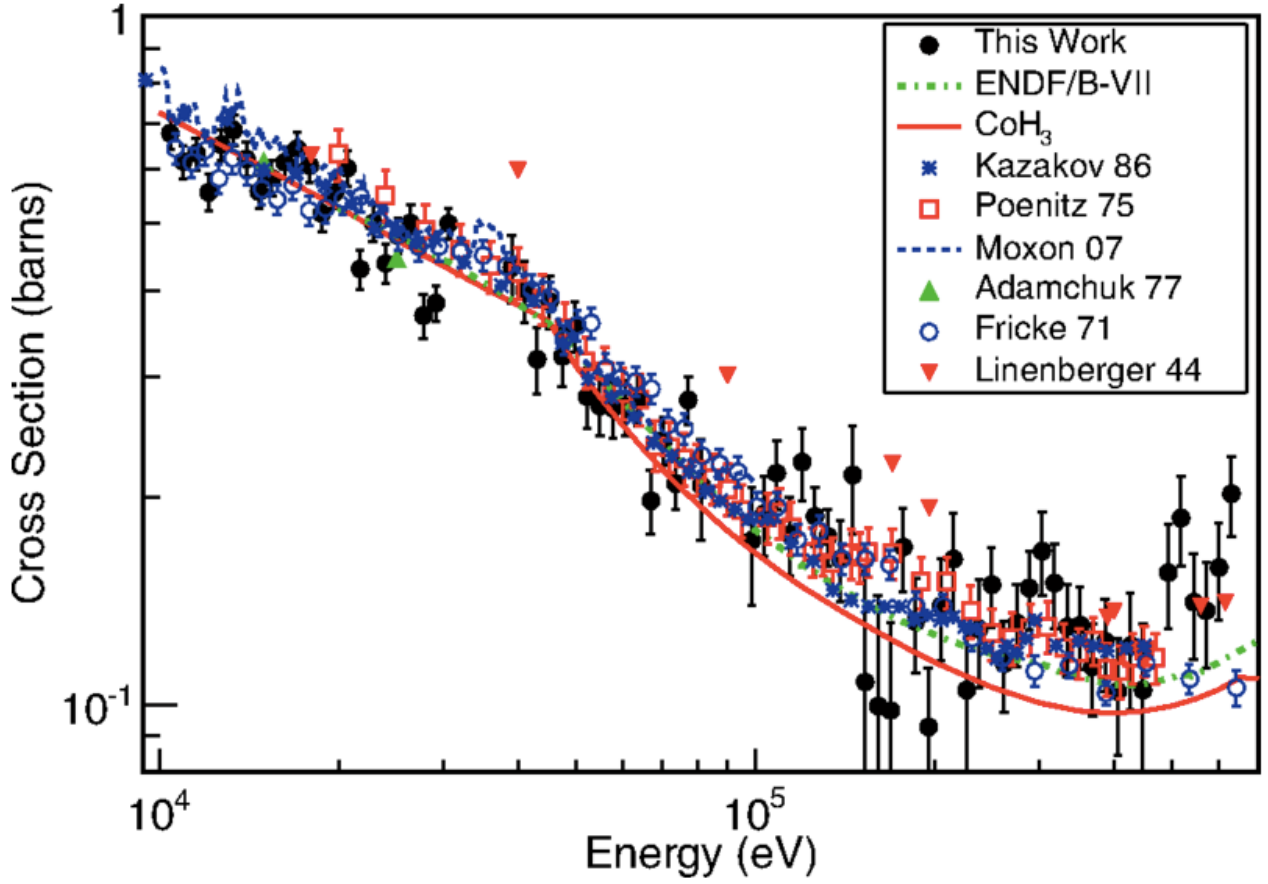


U-238 n,gamma



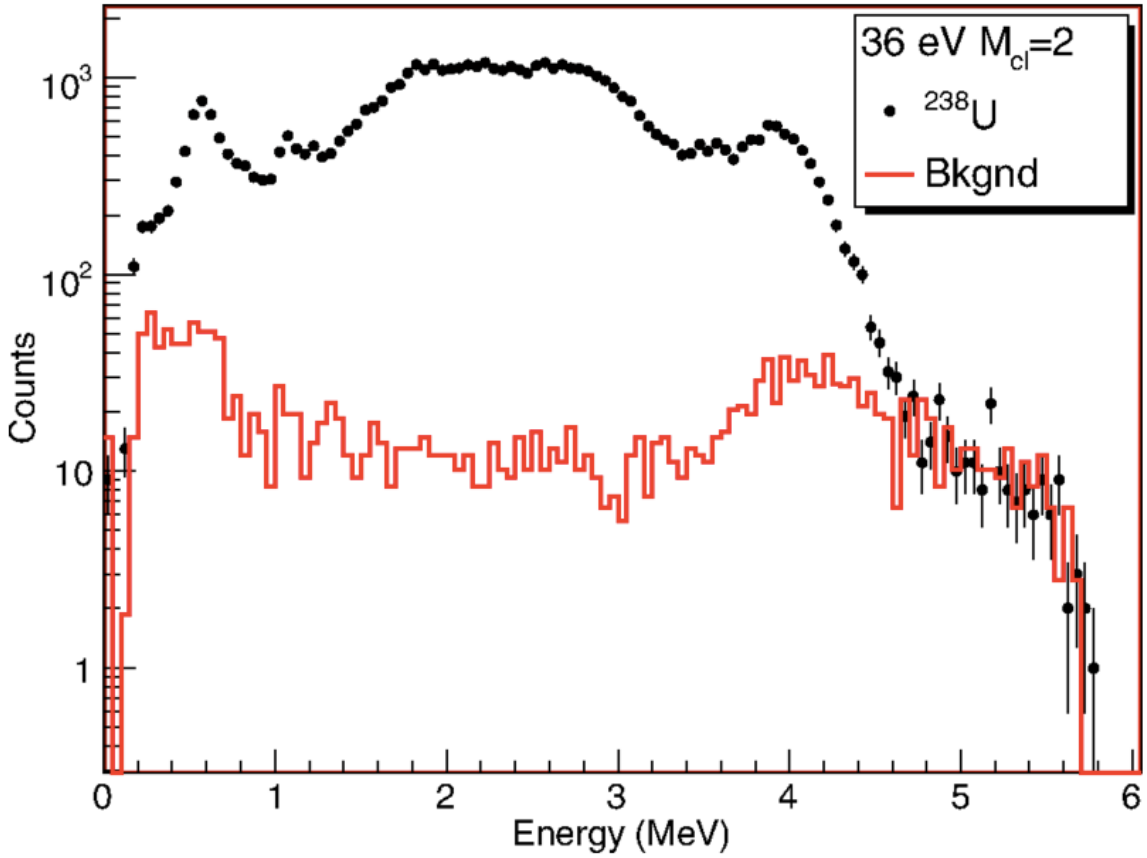
Measured $^{238}\mathrm{U}(n,\gamma)$ cross section compared to previous measurements by Moxon [31], Adamchuk [32], Fricke [30] and Linenberger [33].

U-238 Unresolved Region



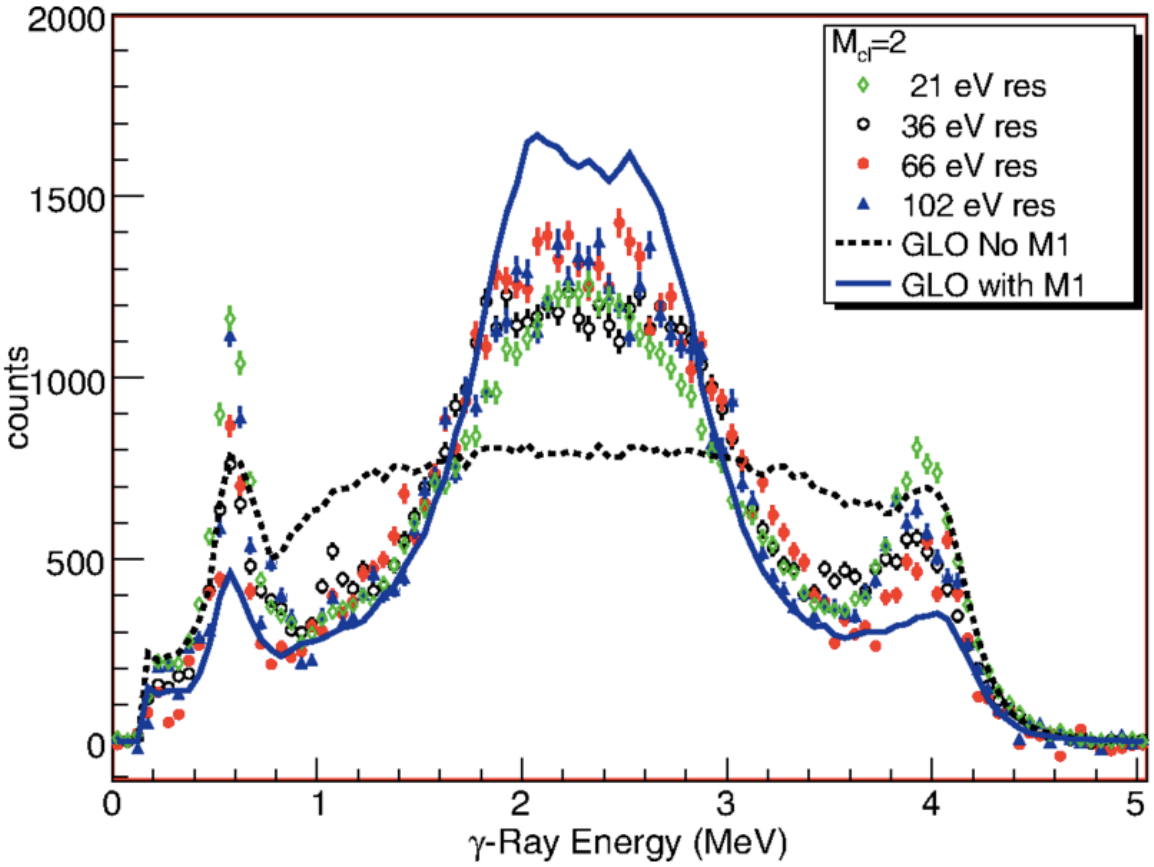
Measured $^{238}U(n,\gamma)$ cross section compared to previous measurements by Kazakov [34], Poenitz [35], Moxon [31], Adamchuk [32], Fricke [30], and

U-238 Gamma Ray Spectrum on a Single Resonance



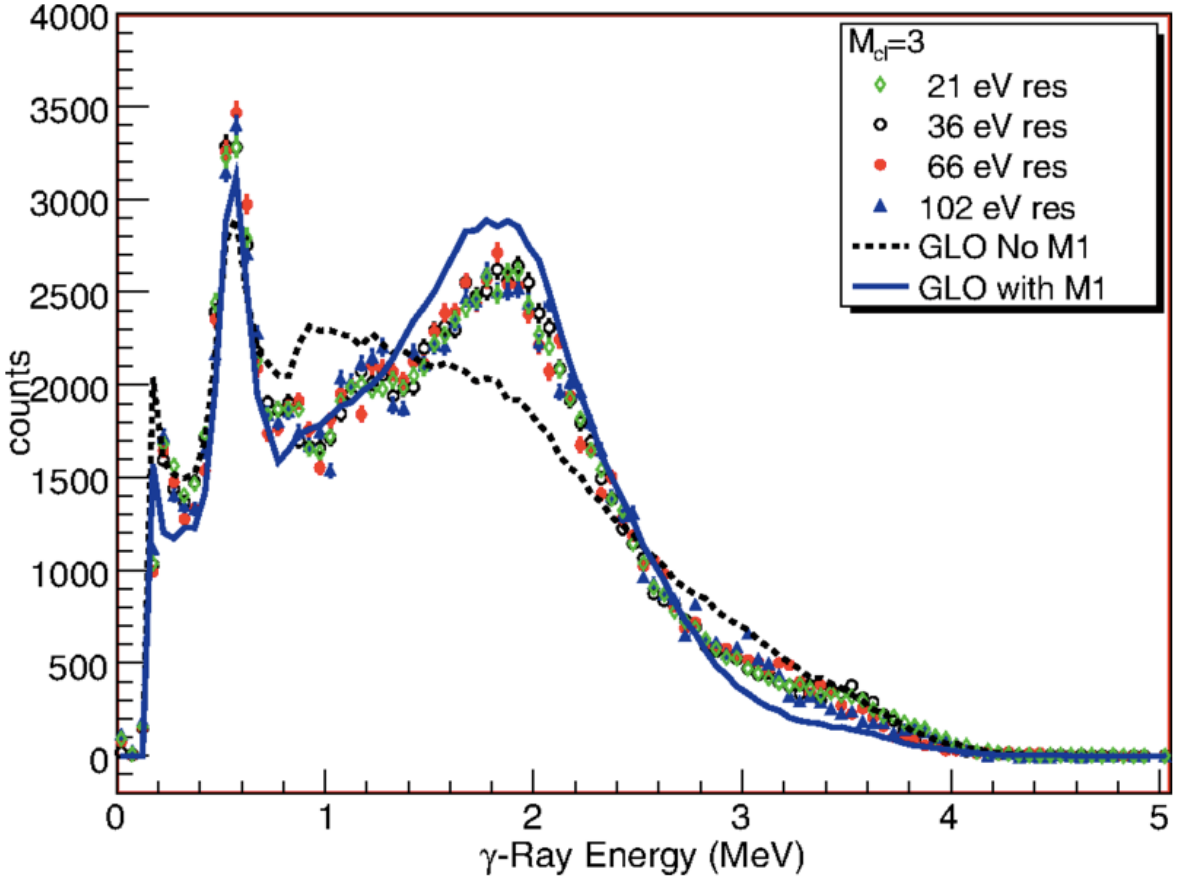
Spectrum of γ rays with $M_{cl} = 2$ from the decay of the 36 eV resonance in ^{238}U(n,γ) compared to the blank background subject to the same data reduction gates.

Comparison to DICEBOX



Spectrum of γ rays from the decay of the four resonances in $^{238}\mathrm{U}(n,\gamma)$, $M_{cl} = 2$, compared with DICEBOX calculations (GLO) with parameters as described in the text.

DICEBOX Multiplicity 3



Spectrum of γ rays from the decay of resonances in $^{238}\mathrm{U}(n,\gamma)$, $M_{cl} = 3$, compared DICEBOX calculations.

U-238 Study Conclusion

- The gamma ray spectra observed required M1 transitions in the DICEBOX cascades.
- The M1 transitions are due to scissor-mode vibrations as opposed to E2 rotations

ENDFB-VI Problem Solved

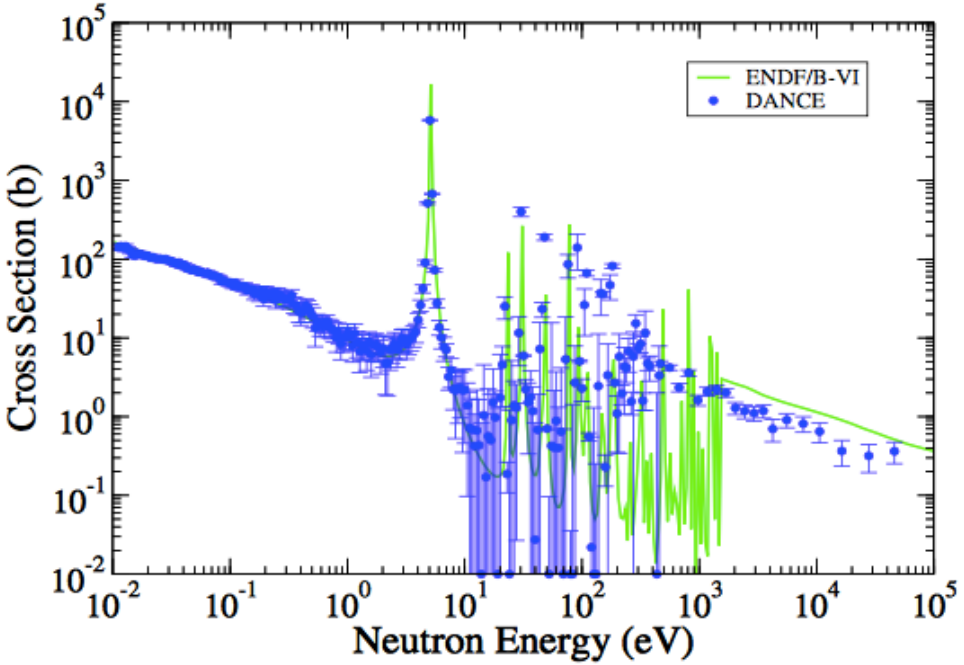


FIGURE 5. ^{234}U capture cross section measured at DANCE. The data is normalized to the thermal neutron capture cross section.